phys. stat. sol. (b) 220, 483 (2000)

Subject classification: 74.50.+r; 74.72.Hs; 74.76.Bz; S10.15

Structural and Electrical Properties of Grain Boundary Josephson Junctions Based on Bi₂Sr₂CaCu₂O_{8+δ} Thin Films

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(Received November 1, 1999)

An in situ deposition sputtering process at high pressure has been developed for preparing high quality superconducting $Bi_2Sr_2CaCu_2O_{8+\delta}$ thin films on different substrates. Both microstructural and electrical properties were well characterized by TEM, AFM, RBS, X-ray diffraction, resistivity and magnetic susceptibility. The high reproducibility of the film quality facilitated a detailed study of Josephson effect in bicrystalline grain boundary junctions (GBJs). Thin films were deposited on (001) SrTiO₃ bicrystals with misorientation angles of 24° and patterned by a photolithography process using Br-ethanol chemical etching. The width of the microbridges ranges from 10 to 50 μ m. The critical current densities across the grain boundary have been measured and compared to the critical current in the film. A modulation in the critical current was found under magnetic field and also Shapiro steps in the I-V curves under microwave irradiation have been observed indicating a Josephson behavior. Electrical properties are well described by the resistively shunted junction (RSJ) model. The I_cR_n product reaches values around 2.0 mV at 4.2 K.

Introduction Since the discovery of high- T_c superconductors there is a rising interest in Josephson junctions made of these materials for applications in cryoelectronics. The main interest focuses on the materials $Y_1Ba_2Cu_3O_{7-\delta}$ (YBCO) and $Bi_2Sr_2Ca_1Cu_2O_{8+\delta}$ (BSCCO) because they exhibit a high transition temperature, and due to the fact that good quality thin films can be deposited by various techniques. Despite a few disadvantages possessed by YBCO, such as the poor stability of the oxygen stoichiometry and fast degradation in air, this material system is mainly used today because of the relative ease of preparation. The bismuth-based superconductor is more difficult to deposit, although the same methods have been employed as for YBCO, i.e., metalorganic chemical vapor deposition, laser ablation, molecular beam epitaxy, and dc sputtering. With each method, thin BSCCO films can be deposited with nearly the right stoichiometry. However, there are very few publications, which report a transition temperature near the optimum value of 94 K, which has been reported for single crystals [1].

In contrast to YBCO there is only little knowledge on the superconducting properties of grain boundaries in the other cuprate superconductors. The study of individual grain boundaries in BSCCO and other oxide superconductors is highly important in order to further clarify the physical origin of the weak link nature of grain boundaries. Also it is important to know if BSCCO bicrystal junctions are useful for cryoelectronic devices.

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484 W. Lopera et al.

In this work we report the structural and electrical properties of bicrystal grain boundary Josephson junctions based on high quality epitaxial thin films of $Bi_2Sr_2CaCu_2O_{8+\delta}$.

Experimental For the fabrication of BSCCO grain boundary Josephson junctions, initially, BSCCO thin films were deposited by a high-pressure dc sputtering technique [2]. We used pure oxygen at 3.5 mbar of pressure as sputtering gas. The deposition temperatures ranged between 880 and 930 °C. BSCCO films were patterned by UV photolithography and Br-ethanol etching to produce 10 to 50 μm wide microbridges across the grain boundary. The thickness of the Bi-2212 films were between 100 and 300 nm and were controlled using the deposition times and the sputtering rates. Typical deposition rates were 1000 Å/h. The films were annealed for 30 min after the deposition keeping the before mentioned parameters for oxygen pressure and temperatures. In addition, BSCCO films were cooled down to 700 °C and post-annealed for 45 min at 0.01 mbar oxygen pressure. The substrates used were single crystals and bicrystals of SrTiO₃.

The fabricated films were almost single phase and were fairly well oriented with the c-axis mainly oriented perpendicular to the substrate surface, as exhibited by different techniques. In θ -2 θ scan X-ray measurements, only (00l) peaks were observed, indicating a strong c-axis orientation. The c lattice parameter calculated from (00l) reflections was 30.929 Å. The spread of the distribution of c-axis oriented grains tilted away from the surface normal was obtained from the rocking curves. The films showed rocking curves around the (00 $\underline{10}$) diffraction peak with full width at half-maximum (FWHM) between 0.2° and 0.3° , indicating good crystalline quality of the samples.

Full epitaxial growth requires not only c-axis alignment but also alignment in the abplane. Thus we have used Rutherford backscattering spectrometry (RBS) combined with channeling measurements to verify the composition and epitaxial quality of the layers (see Fig. 1). RBS spectra and channeling data for BSCCO films showed a minimum yield of about 18%, indicating good epitaxial growth of the layers.

In order to analyze the microstructure and the interfaces between the different layers, cross-sectional high-resolution transmission electron microscopy (HRTEM) analysis was carried out using a Jeol 4000 EX electron microscope. A TEM picture of the

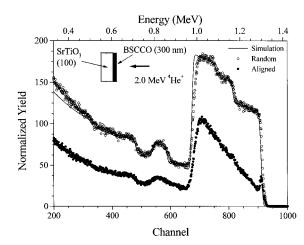


Fig. 1. Rutherford backscattering spectrometry (RBS) combined with channeling for a $Bi_2Sr_2CaCu_2O_{8+\delta}$ thin film deposited on $SrTiO_3$. The spectra show a good epitaxial growth of the film

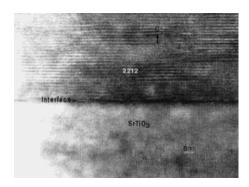
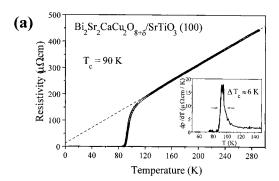


Fig. 2. Image of the interface between the BSCCO (2212 phase) film and the $SrTiO_3$ substrate by transmission electron microscopy. The film exhibits homogeneous epitaxial growth with c-axis normal to the substrate

interface between a BSCCO film (2212 phase) and the substrate is presented in Fig. 2. As reported in the figure, the interface is abrupt and clean, and the film exhibits homogeneous epitaxial growth with c-axis normal to the substrate. A mean roughness of around 4.3 nm in the surface of the films was found by atomic force microscopy (AFM).

Resistivity as function of the temperature for a typical BSCCO thin film is shown in Fig. 3a. The measurements show a good metallic behavior of the film in the normal state and a high critical temperature of around 90 K. Figure 3b shows the real and imaginary parts of the ac susceptibility as function of temperature for the same sample. The diamagnetic onset is in agreement with the critical temperature in resistivity and the sharpness of the transition is lower than 1 K reflecting the high quality of the film.

Resistivity and I-V characteristics were measured using a four-probe technique.



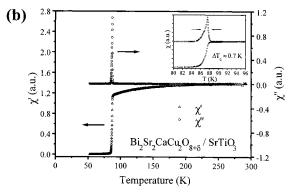


Fig. 3. a) Resistivity and b) susceptibility (real and imaginary parts) as function of temperature for a typical BSCCO thin film. The film shows a good metallic behavior in the normal state and a high diamagnetic onset of 90 K. The sharpness of the transition reflects the high quality of the film

486 W. Lopera et al.

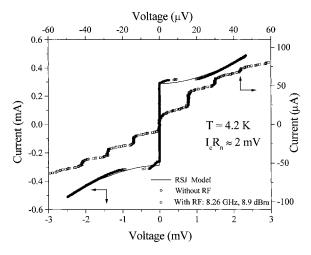


Fig. 4. I-V curve of a 10 μ m wide Bi₂Sr₂CaCu₂O_{8+ δ} bicrystal junction at 4.2 K. The solid line shows the fitting with the RSJ-model and the squares correspond to the I-V measurements under microwave irradiation of 8.26 GHz and 8.9 dBm. Shapiro steps can be observed

Results and Discussion The current-voltage characteristics (IVCs) of a typical 10 μm wide BSCCO bicrystal junction with a missorientation angle of 24° are shown in Fig. 4 (circles). The IVCs follow the shape of the resistively shunted junction (RSJ) model [3]. The I_cR_n product, where I_c is the critical current and R_n is the resistance of the junction in the normal state, is around 2.0 mV. This value is comparable with similar bicrystal junctions of YBCO [4] and higher than values obtained in former publications for BSCCO bicrystal junctions [5]. The critical current density, J_c , for this junction was 1.3×10^4 A/cm², and for all the junctions J_c was always about two orders of magnitude lower than J_c in BSCCO films. The squares in Fig. 4 correspond to the I-V measurements under microwave irradiation of 8.26 GHz frequency and 8.9 dBm power. Shapiro steps at fixed voltages were induced by the microwave irradiation. The separation between these steps is around 17 µV and it is in agreement with the value predicted by the Josephson theory: hf/2e, where f is the frequency of the microwave [6]. The dependence of the Shapiro steps with the microwave power is shown in Fig. 5. Steps up to the 5th order can be observed. The amplitude of the step size is reduced with increasing rf power, but a modulation of the step size with increasing power as predicted by the Josephson effect [6] was not observed for the measured values of power.

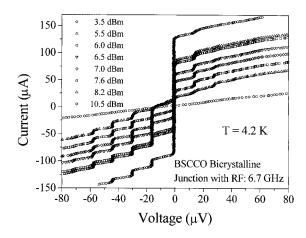


Fig. 5. Series of *I–V* curves with increasing microwave power measured at 4.2 K for a BSCCO Josephson junction. Shapiro steps up to the 5th order can be observed

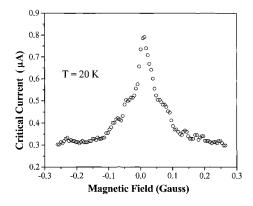


Fig. 6. Magnetic field dependence of the critical current for a BSCCO Josephson junction at 20 K. A modulation of the critical current with the magnetic field is observed

Figure 6 shows the magnetic field dependence of the critical current, $I_c(B)$, of a 30 μ m wide BSCCO bicrystal junction with 24° *ab*-tilt at 20 K. A clear Fraunhofer pattern is not visible as expected for very small junction size, because the width W of the junction is large in comparison with λ_I

 $(W > 4\lambda_{\rm J})$ [7]. Here $\lambda_{\rm J} = (\Phi_0/4\pi\mu_0J_c\lambda_{\rm L})^{1/2}$ is the Josephson penetration depth [6]; Φ_0 is the magnetic flux quantum, $J_{\rm c}$ the critical current density across the grain boundary, μ_0 the vacuum permeability, and $\lambda_{\rm L}$ the London penetration depth. Nevertheless, a modulation of the critical current with the magnetic field can be observed.

Summary and Conclusions Josephson junctions based on high quality epitaxial thin films of $Bi_2Sr_2CaCu_2O_{8+\delta}$ grown on bicrystal substrates of $SrTiO_3$ have been fabricated successfully. The films have high epitaxial quality as shown by X-ray, RBS and TEM. Electrical and magnetic properties of the films have been studied by resistance and susceptibility measurements as function of temperature, giving T_c around 90 K and good metallic behavior in the normal state. The I-V characteristics of the junctions show a clear RSJ-like behavior with I_cR_n values around 2.0 mV at 4.2 K. Shapiro steps up to the 5th order at different microwave power could be observed. The magnetic field dependence of the critical current was measured and a modulation of I_c with the field was observed. The high values of the I_cR_n products and the similar superconducting transport properties of the BSCCO in comparison with YBCO bicrystal junctions suggest an interesting alternative for BSCCO in cryoelectronic devices.

Acknowledgements This work was supported by COLCIENCIAS, Colombia, under contract No. 1106-05-197-95 and by Universidad de Antioquia, CODI project No. IN305CE.

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